Investigation of ozone yield of air fed ozonizer by high pressure homogeneous dielectric barrier discharge

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We succeed in generating an atmospheric and high pressure homogeneous dielectric barrier discharge (DBD) in dry air by using a simple DBD device. So far, we have tried to apply the homogeneous DBD to an ozonizer and found that the ozone yield is higher by the homogeneous discharge mode than by the conventional filamentary discharge mode in larger specific input energy region. In this work, we investigated the effect of gas pressure (from 0.1 MPa to 0.2 MPa) on the ozone yield by homogeneous DBD. The results showed that increase of gas pressure does not improve the ozone yield, because the reduced electric field strength in high pressure homogeneous DBD decreased with increasing the gas pressure.

1. Introduction

The dielectric barrier discharge (DBD) is composed of many filamentary micro-discharges (FDs), and it can be applied to ozone generation [1], gaseous pollution control [2], air flow control [3] and so on. Regarding ozone generation and gaseous pollution control, various methods have been studied to improve the energy efficiency. In our laboratory, efficient oxidation methods of NO in diesel exhaust gas by DBD have been investigated [4]. One of the investigations was a numerical simulation of chemical reactions. The conclusion was that the efficiency was governed by the diffusion process of O radicals and ozone, which were generated in the very thin FD column [5]. Therefore, a homogeneous DBD was expected to improve efficiency of ozone formation, NOx reduction and so on.

In 2009, we succeed in generating an atmospheric pressure Townsend discharge (APTD) in air by a simple DBD device using alumina barriers (Supplier: Kyocera Corporation, purity of Al_2O_3: 92%) and plane electrodes [6], [7]. This discharge is homogeneous without any FDs in a discharge gap. So far, we have investigated the difference of ozone generating efficiencies by FD and APTD [8]. The results showed that the efficiency was higher by the FD mode than by the APTD mode in smaller specific input energy region. However in the region that the specific input energy is larger than 420 J/L, the APTD mode showed higher efficiency than FD mode. In the literature [9], an operation under short discharge gap and optimized gas pressure is advantageous for efficient ozone generation from air.

In this paper, we investigated the ozone generation characteristics of the Townsend discharge (TD) type air fed ozonizer at higher gas pressure.

2. Experimental setup

2.1. High pressure ozone generator

Fig. 1 shows experimental setup. This system consists of a H.V. power source, measurement devices of electrical characteristics and ozone concentration, a chamber, a DBD device, a digital camera and an image intensifier. Dry air (absolute humidity: 119.3 mg/m^3) was used as source gas of ozone generation. The flow rate was fixed to 4.0 L/min (25 °C, 1013 Pa) using a mass flow controller (Horiba, SEC-400mk3). The gas pressure in the chamber was changed from 0.1 MPa to 0.2 MPa (absolute pressure). Ozone concentration was measured by an ozone monitor (Ebara Jitsugyo, EG-3000B/01).

AC high voltage was applied to the DBD device by a step-up transformer. The maximum applied voltage and frequency were 24 kVp (zero-to-peak voltage) and 600 Hz respectively. The applied voltage and the current were measured by an oscilloscope (Tektronix, TDS-2024B, 200 MHz, 2.0 GS/s) using a H.V. probe (Pulse Electronic Engineering, EP-50K, 2000:1) and a differential probe (Yokogawa Electric Corporation, 700924, 100 MHz) respectively. An integral of the current (charge q) was measured from the voltage drop across an integral capacitor. Besides, the discharge power was calculated by multiplying the area of V–q Lissajous figure by power frequency. Discharge photographs were taken by a digital camera (Nikon, D200) with an image intensifier (Hamamatsu Photonics, C5100)

2.2. DBD device

Fig. 2 shows a DBD device. The gap length was fixed to 2.1 mm using spacers. The barrier material is alumina (Kyocera Corporation, Type: A473). The

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14. ABSTRACT
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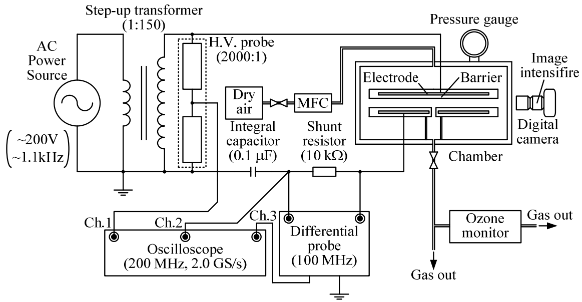
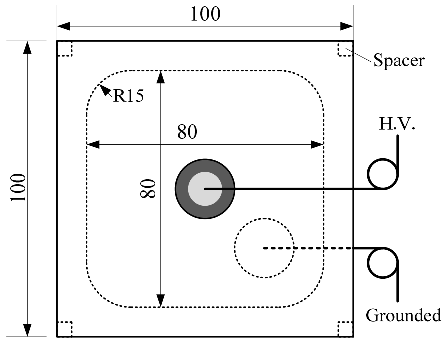
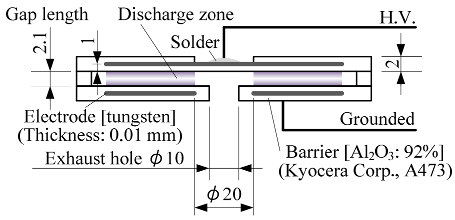


Fig. 1. High pressure ozone generator.



(a) Top view



(b) Side view

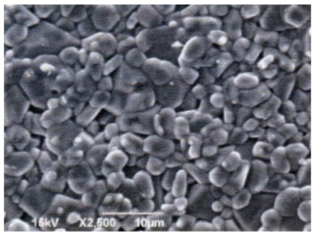
Fig. 2. Barrier discharge device.

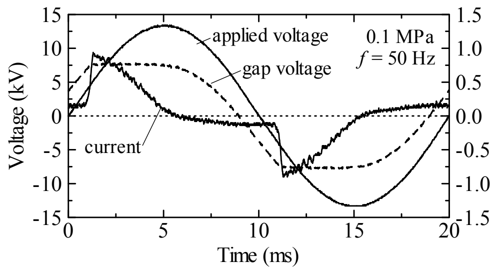
size and thickness are 100 cm^2 and 2 mm respectively. The electrode material is tungsten, and its effective area and thickness are 58.9 cm^2 and 0.01 mm respectively. The electrode was implanted into alumina barrier in order to avoid generation of abnormal discharges from edges of electrodes. Therefore, the barrier thickness from the tungsten film electrode surface and the barrier surface is 1 mm. Physical properties of the barrier material are shown in Table 1.

2.3. Discharge mode

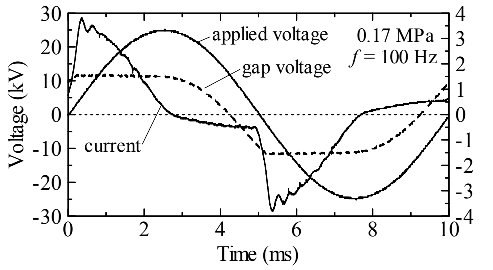
Fig. 3(a) and (b) show waveforms of the applied voltage, gap voltage and current at 0.1 MPa and 0.17 MPa. In case of 0.1 MPa and 50 Hz, the discharge started at around 2 ms and 12 ms in this figure, and during the discharge period, the current was continuous without any pulse. Once a discharge generated in the gap, the gap voltage was maintained to almost a constant value of 8.5 kV. On the other hand, in case of 0.17 MPa and 100 Hz, the discharge started at around 0 ms and 5 ms, and the current flowed continuously as same as the case of 0.1 MPa.

Table 1 barrier material

Material	Al_2O_3
Purity	92%
ε (1 MHz)	9.1
tan δ (1 MHz)	5×10^{-4}
Surface roughness Ra	0.390 μm
SEM image (×2,500)	



(a) 0.1 MPa

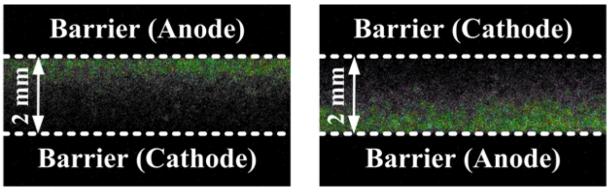


(b) 0.17 MPa

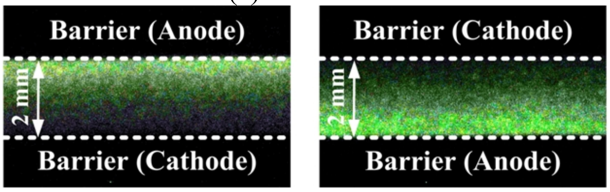
Fig. 3. Applied voltage, gap voltage and current.

The gap voltage became higher, and it was maintained to almost a constant value of 11.4 kV. These current waveforms were completely different from the gathering of high pulse currents observed in typical DBD.

Fig. 4(a) and (b) show the discharge photographs, which were taken by the digital camera with the image intensifier. In case of 0.1 MPa, FDs were not recognized and the luminosity gradually increased from cathode to anode. A layer with strong luminosity appeared near the barrier surface of the anode side. In case of 0.17 MPa, FDs were not recognized and the luminosity gradually increased from cathode to anode as well. These appearances indicate that primary and secondary electrons drift uniformly to the alumina barrier over the anode, and they ionize the gas and generate electron avalanches [10].



(a) 0.1 MPa



(b) 0.17 MPa

Fig. 4. Discharge photograph.

3. Ozone generating characteristics

Ozone generating experiments were carried out using the high pressure ozone generator at various gas pressures. The ozone concentration and the ozone yield were plotted against a specific input energy (SIE). Here, the specific input energy is the energy input to a unit gas volume, namely it is the ratio of discharge power to a flow rate.

Fig. 5 shows the ozone concentrations as a function of SIE for different gas pressure. All of the ozone concentrations increased with increasing SIE. When we check the data, for example, at the SIE of 300 J/L, ozone concentration is 940 ppm at 0.2 MPa but it becomes 1086 ppm at 0.1 MPa.

Fig. 6 shows the ozone yields as a function of SIE for different gas pressure. In this figure, we found that the highest ozone yield was obtained at gas pressure of 0.1 MPa, and all of the ozone yields decreased slightly with increasing SIE. We also found that, in case of 0.2 MPa, the decrease of ozone yield was very slight.

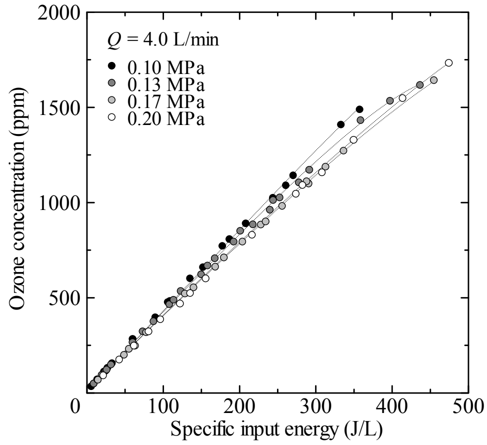


Fig. 5. Ozone concentration as a function of SIE at various gas pressures.

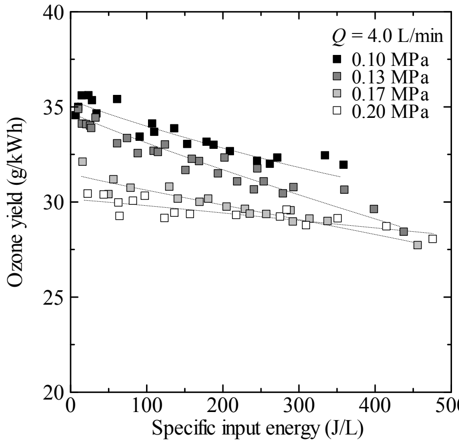


Fig. 6. Ozone yield as a function of SIE at various gas pressures.

4. Discussions

Firstly, we discuss why ozone yield decreased slightly with increasing SIE. In order to clarify this reason, we measured the barrier surface temperature of grounded electrode side. As the result, in case of 0.1 MPa, the temperature was found to be higher than 100 °C at higher specific input energy region. Generally, the thermal decomposition of ozone becomes active in the gas temperature of above 100 °C. Therefore, we are now thinking that the barrier surface temperature seems to be a cause of slight decrease of ozone yield.

Next, we discuss why the highest ozone yield was obtained at gas pressure of 0.1 MPa. In a plasma zone, atomic oxygen is generated by electron impact (R1), and ozone is formed by three body reaction (R2).

$$e + O_2 \rightarrow e + 2O \qquad \text{(R1)}$$

$$O + O_2 + M \rightarrow O_3 + M \qquad \text{(R2)}$$

where, M is a third body collision partner (O, O_2, and/or N_2). From these reactions, it is understandable that high electron energy enhances the dissociation of O_2, and thus the generation of ozone will increase. Next, we checked changes of the discharge sustaining voltage (V_s) and reduced electric field strength (E/n).

Fig. 7 shows the discharge sustaining voltage (V_s) at various gas pressures. Here, the discharge sustaining voltages were determined by measuring a V–q Lissajous figure [11]. The discharge sustaining voltage increased with increasing gas pressure. Next we calculated the E/n (Td) [11].

Fig. 8 shows the reduced electric field strength at various gas pressures. The highest reduced electric field strength was obtained at 0.1 MPa.

In this experiment, the flow rate was fixed to 4.0 L/min using the mass flow controller. Therefore, the gas velocity becomes the minimum at gas pressure of 0.2 MPa, because the flow rate was a converted value at 25 °C and 1013 Pa. The smaller gas velocity is not efficient for the cooling of barriers and thereby thermal decomposition of ozone increased.

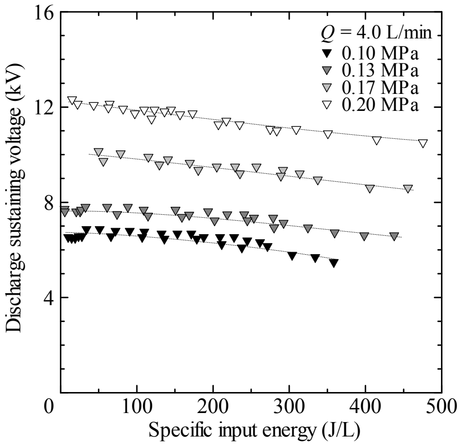


Fig. 7. Discharge sustaining voltage as a function of SIE at various gas pressures.

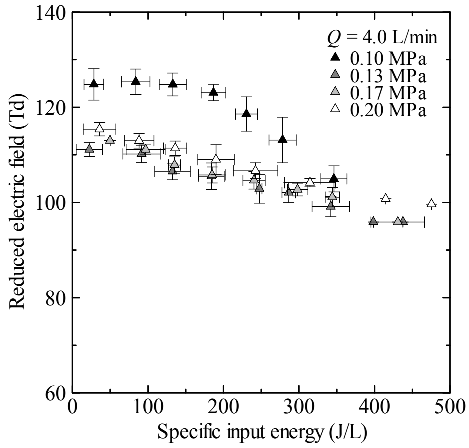


Fig. 8. Reduced electric field strength as a function of SIE at various gas pressures.

In summary, it is apparent that the highest ozone yield of TD type air fed ozonizer was obtained at 0.1 MPa, because a higher electron energy can be obtained in a higher reduced electric field strength and a higher cooling effect of barriers can be also obtained in a low gas pressure. Therefore, the lower gas pressure is better for the dissociation of O_2 and thus good for ozone generation.

Finally, we discuss why lower ozone yield was obtained at 0.2 MPa, and why the decrease of ozone yield was very slight. We are now thinking that the reactions (R1) and (R2), and discharge poisoning by NOx (especially NO and NO_2) [1] were suppressed by the low reduced electric field strength in the discharge. In order to clarify this phenomenon, further study on the gas analysis within the ozone gas by an FTIR spectrometer with a long path gas cell is necessary.

5. Conclusions

We set up the Townsend discharge type air fed ozonizer to investigate the effect of gas pressure (between 0.1 MPa and 0.2 MPa) on ozone generation characteristics. The experimental results obtained are as follows;

(1) The ozone concentrations increased with increasing SIE. When we check the data at a SIE of 300 J/L, ozone concentration of 940 ppm increased to 1086 ppm with decreasing the gas pressure from 0.2 MPa to 0.1 MPa.

(2) The highest ozone yield was obtained at gas pressure of 0.1 MPa, and all of the ozone yields decreased slightly with decreasing SIE.

(3) The highest reduced electric field strength was obtained at the gas pressure of 0.1 MPa, and this is a reason that the ozone yield was the maximum at 0.1 MPa.

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